

## **REMARKS**

Reconsideration and withdrawal of the examiner's rejections under 35 USC §§ 112, 102 and 103 is respectfully requested in view of the above amendments and the following remarks. The applicant would like to thank the examiner for her time and kind cooperation in this matter.

### ***35 USC § 112***

In response to the examiners § 112 rejections, the claim 5 rejection has been obviated by the cancellation of the clause. It is respectfully submitted that regarding claims 6, 8 and 9 and the 'amine', 'imine' confusion; iminodiacetic acid also known as deglycine, i.e.,  $\text{HOOCCH}_2\text{NHCH}_2\text{COOH}$ , is an amine. The diagram on page 7 of the instant specification, shows iminodiacetic acid (top left), which is an amine-containing diacid. The name iminodiacetic acid is a peculiarity of the chemical naming system, but doesn't mean that it contains an 'imine'. Consequently, a person skilled in the art would understand the claim scope.

### ***35 USC §§ 102 and 103***

The Examiner has made the following rejections: Claims 1-4 under 35 U.S.C. 102(b) as anticipated by Makhoul et al (US 3,686,111); Claims 6 and 8 under 35 U.S.C. 102(e) as anticipated by Pereira et al (US 2003/0199593); Claim 7 under 35 U.S.C. 102(e), alternatively 103(a), as anticipated by Pereira et al (US 2003/0199593), or obvious over + Encl. Polymer Sci. and Tech. (vol. 11, page 44); Claims 1-5 under 35 U.S.C. 103(a) over Carswell et al (WO 01/63037) in view of Letton (US 4,260,529); and, Claim 9 under 35 U.S.C. 103(a) over Marans et al (US 3,515,747) in view of Pereira (US 2003/0199593).

Applicant's respectfully traverse each of the examiner rejections as described above. However, in order to expedite examination of the instant case, claim 1 has been amended to a composition instead of the polymer itself (incorporating the limitations of claims 4 and 5) and further to specify the monomers which are used to make up the polyester.

Claim 4 has been amended; support is page 5, lines 11-13 and claim 5.

Claim 2 has been amended to specify that the method is directed to cellulosic textiles. Support is original claim 3 and examples 2 and 4 which are cotton fabrics; and further amended in the same way as claim 4 to describe the polyester.

Claims 1, 3 and 5 are cancelled as being redundant.

Claim 8 has been amended to clarify that the diacid is the amine containing material. Support is at page 5, lines 27-28.

***Makhouf, et al. (U.S. 3,686,111)***

Makhouf discloses a non-aqueous polymeric pseudo dispersion, for dispersing pigments. The dispersion comprises a) particles of acrylic polymer in aliphatic hydrocarbon, b) active solvent for said polymer, c) imine-modified polymerized ethylenically unsaturated carboxylic acid, and d) a dispersion stabilizer (col. 2 line 21 onwards).

The imine-modified polymerized ethylenically unsaturated carboxylic acid (c) may be functionalised to become azetidinium (col. 4, line 53), while the dispersion stabilizer (d) can be a polyester (col. 5, line 14 onwards, especially line 40). Applicant's respectfully submit that the Examiner has thus combined a feature of one component (the imine-modified polymerized ethylenically unsaturated carboxylic acid) with another distinct component (the dispersion stabilizer). The azetidinium functionalised polyester is therefore not disclosed by Makhouf, et al., and a proper prima facie case is absent.

Makhouf, et al., further does not disclose or teach a composition comprising an azetidinium functionalised polyester synthesised by reacting an amine-containing diacid or diol with a suitable co-reactant., and a substrate compatible carrier comprising one or more surfactants as now claimed.

The Examiner asserts that this reference shows that the method of preparing a polyester of original claim 1 (an azetidinium functionalised polyester) is known. However, applicant's respectfully submit this is an incorrect description of the disclosure of Pereira.

To disclose the subject matter of claim 6, Pereira would have to disclose the claimed method of making an azetidinium functionalised polyester, following the method steps a) and b). It is respectfully submitted that Pereira, however, does not disclose the formation of an azetidinium functionalised polyester, nor does it disclose step b) of claim 6.

Regarding step b), the Examiner specifically relies on [0073] on page 8 of Pereira to show that the step of reacting a polyester with an epichlorohydrin is disclosed, however, this paragraph in fact teaches the reaction of polyester with the reaction product of epichlorohydrin and bisulfite (0073, line 12). The bisulfite reacts with the epoxide of the epichlorohydrin to open the ring (see J. Org. Chem., Vol 40, No. 3, 1975, enclosed). This product (epichlorohydrin has a chlorine substituted for a hydrogen group on the CH<sub>3</sub> moiety in formula (2)) is what is reacted with the amine in the polyester in para. [0073] of Pereira. This product can no longer be considered as epichlorohydrin as it does not have an epoxide group. Claim 6 is thus novel over Pereira as step b) is not disclosed as the polyester is not reacted with an epihalohydrin for the above reasons.

Furthermore, it is respectfully submitted that the product of the reaction between the epichlorohydrin/bisulfite compound and the amine present in the polyester would not form an azetidinium. To form the azetidinium ring, the nitrogen must react twice with the epichlorohydrin, once to open the epoxide ring, second to react at the CH<sub>2</sub>Cl center, thus forming the 4-membered ring seen on page 7 of the instant application. If bisulfite has already reacted with epichlorohydrin to open the epoxide ring, then it follows that the amine cannot form an azetidinium group as the nitrogen cannot now react twice. Therefore, claim 6 is not anticipated by Pereira. As claims 7 and 8 are dependent on claim 6, then they are also not anticipated by Pereira.

Although the Encyclopaedia reference may teach the additional subject matter of claim 7, it does not teach the missing features of claim 6. Thus the combination of Pereira and the Encyclopaedia reference fails to recite all claim features and so cannot be the basis for a proper prima facie case under section 103.

Applicants further respectfully submit the combination of Pereira with Marans for the 103(a) rejection of claim 9 is not proper, because as claim 9 is dependent on claim 6, and Pereira already has been shown to not disclose all claim limitations of claim 6, then the combination of Marans with Pereira will not teach all claim limitations for claim 9 and form a proper prima facie case

***Carswell et al (WO 01/63037) in view of Letton (US 4,260,529)***

The Examiner relies on Carswell to teach an azetidinium-based surfactant (page 8, line 15) Applicants respectfully assert that this is not a surfactant as asserted by the Examiner, but a polymer. The polymer can be used in fabric treatment compositions. Carswell does not teach that the azetidinium functionalised polymer is a polyester. Instead a polyamide is disclosed.

Letton is relied upon to disclose a cationic biodegradable polyester surfactant. The Examiner then finds it obvious to replace Carswell polyamide with Letton polyester.

This combination does not teach all of the claim features of amended claim 2, as Letton does not teach that the polyester is synthesised by reacting an amine-containing (di)acid or (di)ol with a suitable co-reactant. As all claim features are not recited, the amended claim is unobvious over the combination of references since a proper prima facie case is absent.

Applicants further respectfully submit that although the cationic surfactant of Letton is not a polyester - it may have an ester group (or a ketone, ether, amide etc, col. 3), but it is not a polymer containing ester linkages. The cationic surfactant of Letton is not azetidinium functionalized. Azetidinium is a 4 membered ring with a 4 substituents on the nitrogen. It is not the '5-6 membered ring mentioned by the Examiner. Azetidinium is not mentioned at all in Letton. Thus it is respectfully submitted that the replacement of the polyamide of Carswell with the Letton surfactant would not make an azetidinium functionalised polyester.

Moreover, it is respectfully submitted that the skilled person would not be led to replace a feature of a polymer (Carswell) with a feature of a surfactant (Letton) absent impermissible hindsight. Furthermore, the surfactants of Letton are biodegradable, but this feature is not linked to the ester function, so the skilled person would not be taught that the ester is useful because it is biodegradable.

Finally, it is the amine groups present in the polymer that are reacted to produce the azetidinium function. These amine groups are inherently present in a polyamine, but would not be inherent in a polyester. One would have to choose monomers to make the polyester which additionally contain an amine function according to the instant claims. So a straight replacement of polyamide with polyester would not allow further azetidinium functionalisation, which was found useful for the crosslinking properties with cellulose.

***IDS***

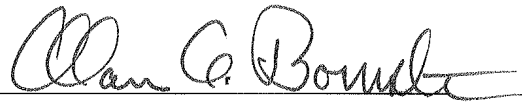
Applicants note that the second page of form 1449 mailed on March 3, 2005, was not returned. It would be appreciated if the examiner could kindly return the signed form with his next response. A copy is enclosed for the examiner's convenience.

### CONCLUSION

In summary, claims 2, 4, 6, 7 and 8 have been amended. Claims 1, 3 and 5 have been cancelled as being redundant. No new matter has been added by these amendments.

In light of the above remarks, applicants submit that the claims now pending in the present application are in condition for allowance. Reconsideration and allowance of the application is respectfully requested. The examiner is invited to contact the undersigned if there are any questions concerning the case.

Respectfully submitted,

A handwritten signature in black ink, appearing to read "Alan A. Bornstein", written over a horizontal line.

Alan A. Bornstein  
Registration No. 40,919  
Attorney for Applicant(s)

AAB/ss  
(201) 894-2180

OTHER DOCUMENTS (Including Author, Title, Date, Pertinent Pages, etc.)

International Search Report, PCT/EP 03/00283, mailed Apr. 24, 2003 – 2 pp.

GB Search Report, GB 0205277.7, dated July 15, 2002 – 1 pp.

CA 107:7751 Abstract of "Makromol. Chem. 1987, 188(4), 791-7, Tezuka, et al."

EXAMINER: Initial if citation considered, whether or not citation is in conformance with MPEP 609; Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to Applicant.

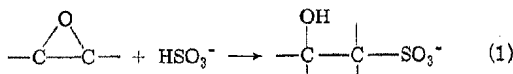
A Kinetic Study of the Reaction between Sulfite Ion and Propylene Oxide<sup>1a</sup>Gerald S. Yoneda,<sup>1b</sup> Michael T. Griffin,<sup>1c</sup> and David W. Carlyle\*<sup>1d</sup>

Department of Chemistry, Texas Tech University, Lubbock, Texas 79409

Received July 8, 1974

The reactive sulfur species in the sulfur(IV)-propylene oxide reaction is sulfite ion. The reaction is first order in sulfite and also first order in propylene oxide under conditions of dilute base in nearly pure water. The reaction order with respect to propylene oxide becomes less than one as the propylene oxide concentration is increased. These observations are consistent with simple nucleophilic attack by the sulfite ion, and with strongly concentration-dependent activity coefficients for propylene oxide.

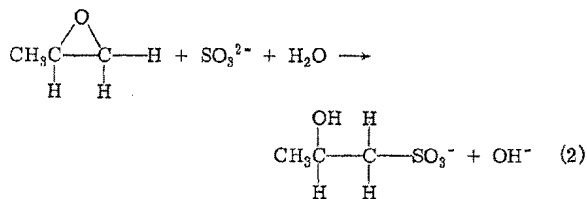
Reactions between epoxides and sulfur(IV) generally result in ring opening<sup>2</sup> as illustrated by eq 1. If the ring car-



bon atoms in the reactant are substituted unsymmetrically, then nonequivalent sites for sulfur(IV) attack exist and either or both of the isomeric products may be obtained.<sup>2</sup> If one of the ring carbon atoms is bound to more bulky substituents than the other, as in propylene oxide, then the major product is usually predicted to have the sulfonate group in the terminal position owing to smaller steric resistance to attack at the terminal carbon atom.<sup>2</sup> However, this generalization is not valid for every reaction. For example, the reactions of sulfite with styrene oxide and with 1,2-epoxyoctane have been reported to give only the corresponding 2-sulfonic acids.<sup>3</sup> The product of the sulfite reaction with propylene oxide, the epoxide of principal interest in the present report, has been reported to give only the terminal sulfonate<sup>4,5</sup> but may give a mixture.<sup>6</sup> Even greater disagreement exists about the product of the sulfur(IV)-epichlorohydrin reaction. A product with a normal terminal sulfonate group has been reported,<sup>7</sup> but the reaction has also been reported<sup>8</sup> to give under some conditions terminal sulfonate by replacement of the chloride, without disturbing the ring. However, this last report has been questioned.<sup>9</sup>

Some very early observations of epoxide reactions with sulfite were made,<sup>10</sup> but not much work has been done recently.<sup>9</sup> One moderately recent study<sup>3</sup> was a stoichiometric and kinetic investigation of the reaction between sulfite and styrene oxide in two phases. It was concluded that sulfite ion is the reactive sulfur(IV) species and that the reactions are first order in sulfite ion; however, an attempt to study the reaction between propylene oxide and sulfite in a single aqueous phase was not successful because the reaction was too fast to study by methods then available.

Owing to our interest in reactions of sulfur(IV)<sup>11</sup> and to the absence of detailed kinetic data for reactions of the class shown in eq 1, we have studied the sulfur(IV)-propylene oxide reaction indicated by eq 2 and have made a few observations of some related reactions.



## Experimental Section

**Reagents.** Oxygen-free aqueous sodium sulfite solutions and sodium perchlorate solutions were prepared as described earlier.<sup>12</sup>

Reagent grade sodium carbonate, sodium hydroxide, perchloric acid, ethylene oxide, propylene oxide, propylene sulfide, epichlorohydrin, epibromohydrin, and deuterium oxide were used without purification. The water used for preparation of each solution was redistilled from laboratory distilled water and was stored in a polyethylene tank.

**Rate and Nmr Measurements.** Rates of reaction 2 were measured at 4.0, 14.1, and 25.0° in aqueous alkaline solutions of 0.25 *M* ionic strength maintained with NaClO<sub>4</sub>. The reactant solutions were contained in 5- or 10-cm spectrophotometer cells positioned in a Beckman ACTA V recording spectrophotometer. The progress of each reaction was observed by measuring the absorbance decrease at 270 nm, where SO<sub>3</sub><sup>2-</sup> is the principal absorbing species. Propylene oxide was in large excess in each experiment. Each individual experiment was first order in sulfite and the pseudo-first-order rate constant was obtained from a plot of ln (*A* - *A*<sub>∞</sub>), vs. time, where *A* and *A*<sub>∞</sub> are the absorbances at 270 nm at a particular time and at completion of the reaction, respectively.

The procedures used in the propylene oxide experiments were also used in attempts to measure the rate of reaction of sulfite ion with ethylene oxide and with propylene sulfide. These same procedures were also used in attempts to measure the rates of reaction with epichlorohydrin and epibromohydrin, except the solvent was 50% ethanol owing to the insolubility of these organic compounds in pure water.

A Varian XL-100 spectrometer was used to measure the proton nmr spectra of the reactants and products of reaction 2 and of the analogous epichlorohydrin reaction in D<sub>2</sub>O solution.

## Results

**Reaction Product.** We used proton nmr spectra to confirm that the sulfonate group in the reaction 2 product is indeed in the terminal position as expected<sup>2</sup> and as asserted by some earlier workers.<sup>4,5</sup> Our measurement of the spectrum of reactant propylene oxide gave an upfield doublet assigned to the methyl group. In addition to the upfield doublet, multiplets centered at 75 and 93 MHz downfield from the doublet and a poorly resolved multiplet centered at 108 MHz downfield from the doublet were observed. The spectrum had the same appearance as spectra reported earlier<sup>13,14</sup> for propylene oxide. In addition, our assignments are in agreement with the earlier ones;<sup>13,14</sup> the methyl group resonance is upfield, the resonances for the two hydrogen atoms on the other terminal carbon atom are at 75 and 93 MHz, and the resonance for the remaining hydrogen atom is at 108 MHz downfield.

Reaction 2 was allowed to occur in D<sub>2</sub>O solution under conditions of excess sulfite so that all the propylene oxide was converted to product. The proton nmr spectrum of the resulting solution had the following downfield resonances (still referred to the upfield doublet for propylene oxide): a doublet centered at 8 MHz assigned to the methyl group, multiplets at 113 and 120 MHz assigned to the hydrogen atoms on the terminal carbon atom containing the sulfonate group, and a multiplet at 190 MHz assigned to the hydrogen atom on the central carbon atom. The assignment of the downfield multiplet is based on the low relative intensity and on the similarity in shape to the analogous mul-

Table I  
Observed Pseudo-First-Order Rate Constants for  
Reaction 2 at 0.25 M Ionic Strength and 0.005–0.01  
M NaOH

$[\text{C}_3\text{H}_5\text{O}],^a$ mol %	$10^3 k',$ $\text{sec}^{-1}$	$[\text{C}_3\text{H}_5\text{O}],^a$ mol %	$10^3 k',$ $\text{sec}^{-1}$
0.534	0.208 <sup>b</sup>	0.192	0.218 <sup>c</sup>
1.08	0.447	0.485	1.49
2.23	1.24	0.980	2.54
3.20	1.44	1.03	2.62
3.44	1.58	2.02	5.64
3.70	1.78	2.06 <sup>d</sup>	5.83
4.74	1.35	2.06 <sup>e</sup>	5.73
6.12	0.940	2.06 <sup>d,f</sup>	5.77
7.58	0.759	4.28	9.90
7.89	0.645	5.75	12.6
0.520	0.510	9.78	15.4
1.05	0.672	17.1	14.2
2.16	2.31	27.3	12.6
4.61	3.08	36.8	11.1
5.95	3.22	41.1	10.6
7.37	2.25		

<sup>a</sup> Mole per cent solvent, considering only propylene oxide and water. <sup>b</sup> Each of the first ten rate constants listed in this column was measured at 4.0°. The remaining six were measured at 14.1°. <sup>c</sup> Each of the rate constants listed in this column was measured at 25.0°. <sup>d</sup>  $[\text{OH}^-]$  was 0.0094 M. <sup>e</sup>  $[\text{OH}^-]$  was 0.00504 M. <sup>f</sup> The reaction occurred in an oxygen-free solution.

Table II  
Observed and Calculated Values for  $k^0$  and Activation  
Parameters for  $k^0$ , Valid in the Limit as  $[\text{PO}]$   
Approaches Zero

Rate constant $k^0$ <sup>a</sup>			$\Delta S^\ddagger$ , eu	$\Delta H^\ddagger$ , kcal/mol
4°	14.1°	25°		
0.045	0.108	0.290	2.5 <sub>5</sub>	14.0

<sup>a</sup> The rate constants are defined by eq 4; the units are (mole fraction)<sup>-1</sup> sec<sup>-1</sup>.

triplet in propylene oxide. We conclude that the hydroxyl group in the reaction 2 product is indeed on the central carbon atom, rather than on the terminal one, because the resonance assigned to this hydrogen atom shifted farther downfield than any of the other peaks, in accord with the greater deshielding strength of OH relative to  $\text{SO}_3^-$  or  $\text{SO}_3\text{H}$ .

We were able to measure an nmr spectrum of epichlorohydrin that agrees with spectra already reported,<sup>16</sup> but the products of the reaction of epichlorohydrin with excess sulfite in  $\text{D}_2\text{O}$  solution apparently include at least two organic species. The nmr spectrum of the product mixture is complex and is not consistent with complete conversion to any one of the three expected products (1-chloro-2-hydroxy-3-sulfonic acid,<sup>7</sup> 1-chloro-3-hydroxy-2-sulfonic acid, or 1,2-epoxy-3-sulfonic acid<sup>8</sup>).

**Phase Properties of the Water-Propylene Oxide System.** The salt-free water-propylene oxide system is known<sup>16</sup> to form two liquid phases at mole fractions of propylene oxide ranging from about 0.17 to about 0.90. The two-phase region exists at all temperatures between the freezing and boiling points of the liquid. The solids that can exist in equilibrium with the liquid phases are pure propylene oxide, pure water, and a clathrate compound propylene oxide · 17 water.<sup>17</sup> Our visual observations of the system show that addition of 0.25 M  $\text{NaClO}_4$  renders the

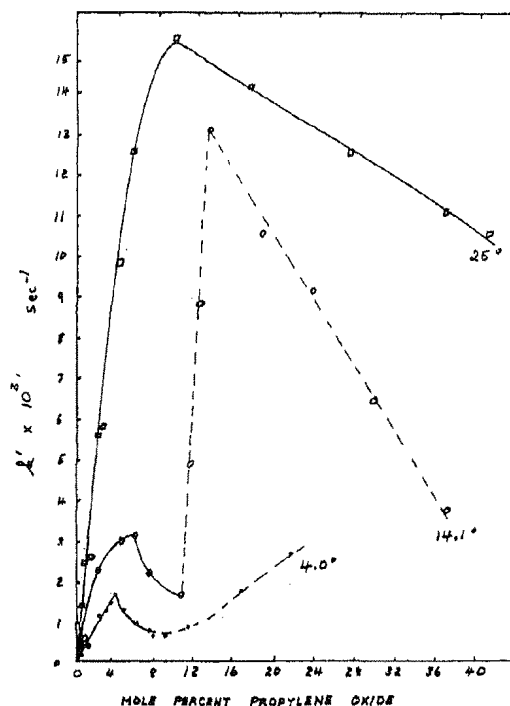


Figure 1. The relationship between observed rate constant for reaction 2, and mole per cent propylene oxide. The dashed lines connect points measured under two-phase conditions.

solvents completely miscible at 25°, but two liquid phases remain at temperatures below about 20°.

**Rate Measurements.** The numerical values of the pseudo-first-order rate constants measured for reaction 2 are listed in Table I. Each of the rate constants listed in the table occurred in a one-phase system and is defined by eq 3. These same rate constants are shown graphically in Fig-

$$-\frac{d \ln [\text{SO}_3^{2-}]}{dt} = k' \quad (3)$$

ure 1, along with some apparent rate constants obtained from two-phase systems.

Experimental results listed in Table I show that small variations in  $[\text{OH}^-]$  do not affect the value of  $k'$ , although experiments in dilute acid solution showed an induction period; reaction was slow until the solution became basic due to production of  $\text{OH}^-$  by reaction 2. Similarly, Table I indicates that deoxygenation of the reaction mixture does not affect the value of  $k'$ . However, solvent phase separation does affect the apparent rate constant, as indicated in Figure 1.

Even the data obtained at 25°, all in a one-phase system, indicate a complex relation between the pseudo-first-order rate constant and the mole per cent<sup>18</sup> propylene oxide at high mole per cent propylene oxide. However, the data obtained at each of the temperatures conform to a first-order dependence on propylene oxide at low  $[\text{PO}]$ .<sup>19</sup> The limiting slopes obtained from Figure 1 lead to the observed rate constants given in Table II, where the rate constant  $k^0$  is defined according to eq 4. The values of  $k^0$  are correlated

$$-\frac{d \ln [\text{SO}_3^{2-}]}{dt} = k^0 [\text{PO}] \quad (4)$$

very well by the absolute rate theory equation, leading to the activation enthalpy and entropy values of 14.0 kcal/mol and 2.5 eu, respectively, for  $k^0$ .

Some attempts were made to measure the rates of sulfite ion reaction with substrates other than propylene oxide.

Table III  
Observed Pseudo-First-Order Rate Constants for  
Reaction of Sulfite Ion with Ethylene Oxide at 0° in  
Alkaline Aqueous Solution

$[C_2H_4O], M$	$10^3 k', \text{sec}^{-1}$
0.091	1.5
0.112	2.3
0.120	2.4
0.188	1.4
0.221	3.9
0.225	3.5
0.296	5.3

The sulfite reaction with propylene sulfide was rapid and yielded a polymeric solid that did not dissolve in any of the polar or nonpolar solvents we tested. Neither epichlorohydrin nor epibromohydrin was sufficiently soluble in water to permit rate measurements in water solution. In addition, both these substrates reacted quite rapidly with sulfite. However, a rate constant was measured for epichlorohydrin at 4° in 50 vol % water-ethanol solution; the rate was first order in sulfite, and based on the assumption that the rate is also first order in epichlorohydrin, the rate constant was about  $0.02 M^{-1} \text{sec}^{-1}$  in a solution initially containing  $4.88 \times 10^{-3} M$  epichlorohydrin,  $0.01 M$  NaOH, and about  $2.30 \times 10^{-3} M$   $SO_3^{2-}$ .

The reaction between sulfite ion and ethylene oxide was also quite fast and study of the reaction was further complicated by the inconvenient volatility of ethylene oxide. The ethylene oxide was added to the spectrophotometer cell by bubbling the gas into the water solution in the cell. The ethylene oxide concentration after completion of the reaction was determined by measuring the absorbance at 260 nm, using  $\epsilon = 1.09$ . The initial portions of the reactions had already occurred by the time rate measurements were started, and the data were only moderately reproducible. The data are listed in Table III, where  $k'$  is defined by eq 3. The  $k'$  data are approximately in accord with eq 5, where  $k = 0.02 M^{-1} \text{sec}^{-1}$ .

$$k' = k[C_2H_4O] \quad (5)$$

### Discussion

The data at low [PO] are consistent with empirical eq 4. A possible minor contributor to the deviation from eq 4 at higher [PO] is a dependence on the activity of water; the activity of water would be expected to remain approximately constant with small changes in [PO] in nearly pure water, but would be expected to decrease slightly as [PO] becomes larger.<sup>16</sup> A probable major contributor to the deviation is the decreasing activity coefficient for propylene oxide with increasing [PO]. Activity coefficients for propylene oxide and water in the salt-free mixed solvent system have been determined at the normal boiling points of the system,<sup>16</sup> but have not been determined at any of the temperatures employed in the present study. The activity coefficient for propylene oxide at the mixed solvent boiling temperatures does decrease sharply in the [PO] range 0–0.4, however, and it appears reasonable to assume similar behavior at lower temperatures in the presence of dissolved salts. We think it is probable that either eq 6 or 7 accurately

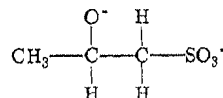
$$-d \ln [SO_3^{2-}]/dt = k a_{PO} a_{H_2O} \quad (6)$$

$$-d \ln [SO_3^{2-}]/dt = k a_{PO} \quad (7)$$

ly represents the rate behavior for reaction 2 in the one-phase reaction mixtures containing up to 0.41 mol fraction

propylene oxide. The first-order dependence on sulfite ion concentration and the negligible importance of terms involving bisulfite concentration are well established by our work. Our conclusion that sulfite is the reactive sulfur(IV) species is in agreement with the conclusion reached from the earlier study,<sup>3</sup> although the earlier study did involve heterogeneous systems and the investigators apparently did not realize that  $SO_3^{2-}$  and  $HSO_3^-$  are in equilibrium in aqueous solutions.

The form of eq 4 is consistent with simple attack of the nucleophile sulfite ion upon the terminal ring carbon atom of propylene oxide to give



as the product of the rate-determining step if eq 7 is correct, or to give the final net reaction product if eq 6 is correct.

With respect to the reactions we studied more briefly than reaction 2, our results with ethylene oxide are not inconsistent with a mechanism analogous to that described just above for propylene oxide. The stoichiometric evidence for the epichlorohydrin reaction permits no generalization except that at least two competing pathways exist for reaction with sulfite. The sulfite-induced polymerization of propylene sulfide is not surprising; if the initial reaction is analogous to reaction 2, then a mercaptan is formed and the mercaptan could act as a nucleophile in attacking another propylene sulfide molecule to give a sulfur bridged dimer containing another mercaptan group capable of continuing the polymerization. A detailed description of an analogous process for the ethoxide-induced polymerization of propylene sulfide has already been presented.<sup>20</sup>

Registry No.— $HSO_3^-$ , 15181-46-1; propylene oxide, 75-56-9; ethylene oxide, 75-21-8.

### References and Notes

- (a) Supported by the Robert A. Welch Foundation; (b) Robert A. Welch Predoctoral Fellow, summer, 1973; (c) Robert A. Welch Undergraduate Scholar, 1972, 1973; (d) address correspondence to this author at R.R. 1, Box 115, Odessa, Missouri 64076.
- J. G. Buchanan and H. Z. Sable, in "Selective Organic Transformations," Vol. 2, B. S. Thyagarajan, Ed., Wiley-Interscience, New York, N.Y., 1972, pp 2–11.
- R. T. E. Schenck and S. Kaizerman, *J. Amer. Chem. Soc.*, **75**, 1636 (1953).
- A. Lambert and J. D. Rose, *J. Chem. Soc.*, 46 (1949).
- J. M. Stewart and H. P. Cordts, *J. Amer. Chem. Soc.*, **74**, 5880 (1952).
- D. P. Sheetz, U.S. Patent 2,914,499; *Chem. Abstr.*, **54**, 20274 (1960).
- S. Tsunoo, *Ber. Deut. Chem. Ges. B*, **68**, 1334 (1935), cited in ref 9.
- W. Schmidt, U.S. Patent 2,265,200; *Chem. Abstr.*, **36**, 1955 (1942).
- E. E. Gilbert, "Sulfonation and Related Reactions," Interscience, New York, N.Y., 1965, pp 161, 162.
- E. Erlenmeyer and L. Darmstaedter, *Z. Chem.*, 342 (1888); L. Darmstaedter, *Justus Liebigs Ann. Chem.*, 148 (1868); F. O. Pazsche, *J. Prakt. Chem.*, **1**, 85 (1870); and L. Brunel, *C. R. Acad. Sci.*, **137**, 63 (1903), each cited in ref 3.
- E. L. Stapp and D. W. Carlyle, *Inorg. Chem.*, **13**, 834 (1974), and references therein.
- D. W. Carlyle, *Inorg. Chem.*, **10**, 761 (1971).
- F. A. Bovey, "Nuclear Magnetic Resonance Spectroscopy," Academic Press, New York, N.Y., 1969, pp 148–150.
- "Sadtler Standard Spectra," Sadtler Research Laboratories, Inc., Philadelphia, Pa., Spectrum No. 10797M.
- Reference 14, spectra 16 and 50Z.
- J. N. Wickert, W. S. Tamplin, and R. L. Shank, *Chem. Eng. Progr. Symp. Ser.*, No. 2, 48, 92 (1952).
- J. C. Rosso and L. Carbonnel, *C. R. Acad. Sci., Ser. C*, **274**, 1868 (1972).
- Our calculation of mole per cent propylene oxide takes into account only water and propylene oxide and does not take solutes into account.
- We are using the symbol [PO] to represent the mole fraction of propylene oxide.
- D. D. Reynolds and D. L. Fields in "Heterocyclic Compounds with Three and Four-Membered Rings," Part One, A. Weissberger, Ed., Interscience, New York, N.Y., 1964, pp 602, 603.